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 Requestor Document Center (is requested to provide the following document)

Date of request ~~02/28/96~~ 6/28/96 Expected receipt of document 1/8/95

Document number none Date of document ~~none~~ ~1970

Title and author (if document is unnumbered)

The Uranium Accountability System &

(This section to be completed by Document Center)

Date request received 7/1/96

Date submitted to ADC 7/11/96

Date submitted to HSA Coordinator 7/1/96

(This section to be completed by HSA Coordinator)

Date submitted to CICO 7/11/96

Date received from CICO 7/16/96

Date submitted to ChemRisk/Shonka and DOE 8/1/96

(This section to be completed by ChemRisk/Shonka Research Associates, Inc.)

Date document received

Signature

THE URANIUM ACCOUNTABILITY SYSTEM

Introduction:

In a gaseous diffusion plant, as with most other continuous production processes, material control is important in evaluating the efficiency of the operation. The material balance for a finite period of time is the instrument from which judgments are made. Components of the balance are the raw material entering the process, the finished material produced, and the worked material in progress. Translating these to the diffusion plant, the raw material is represented by the feed streams; finished material, by the production streams, both top and bottom; and work in process, by the dynamic gas phase inventory. The last component enumerated, i.e., the gas phase inventory, is the most difficult to develop. The reason for this is that the process gas is a heavy, highly corrosive material being measured by pressure and temperatures in the containing equipment. Further, each stage of separation is subdivided into six distinct pressure and temperature regions. The volume of each of these regions is determined by engineering calculation. This means that there are potential bias errors. It is to be noted, however, that there is no known reason to suspect that there are errors of this type in the presently operating equipment.

Superficially, it would appear that a zero material balance is desirable, i.e., all material is completely accounted for. From a practical point of view, this cannot and should not happen on a short term basis. The corrosive quality of the process gas results in a continuous deposition of material on the surfaces of the containing equipment. This represents a loss from the gas phase upon which the short term balances are defined. Over a long period

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Prepared by Union Carbide Corporation-
Nuclear Division, operating contractor for the
U.S. Department of Energy under U.S.
Government Contract No. W-7405-eng-26.

of time in which the equipment is replaced, compensating credit for recovery of deposited material will occur. This suggests that the material balance should reflect a continuous steadystate loss rate which is predictable. To some extent this is true. A complication involved, however, is that statements of gas phase inventory are subject to inherent error. These should be random and will cause fluctuations in the material balance in both directions, viz., apparent gains and losses. The amplitude of these fluctuations is a function of the attention given to instrument calibration, to maintaining proper volume evaluations if changes are made, to proper time differentiation at balance interval interphase points, and to the accuracy with which process data are recorded. Parenthetically, it is to be noted that feed and production streams are measured in cylinders represented by weights, samples, and analyses all of which are controllable by known methods.

It is the intent of the subsequent discussion to delineate the history of the plant with respect to material control, to provide observed experience for that period of time representing current levels of operation, and to define at least a limited program of experimental effort and operational modification to enhance the state of the art.

HISTORY OF THE PROGRAM

Almost coincident with the beginning of operation in the K-25 and K-27 buildings, substantial effort was devoted to the development of inventory estimates and maintenance of material balances. System volumes were calculated from construction drawings. These were subdivided into pressure and temperature regions. Standard instrumentation provided pressures and temperatures for each stage 'B' outlet together with control valve position. Other regional operating data were based upon experimentally probed units or were inferred from "circuit balance" type evaluations. Dynamic inventory data were taken on a daily basis. This included a complete isotopic gradient. The inventories together with the measured feed and withdrawal activity were translated into daily material balances.

During the summer of 1947 with the product assay at about 30% and all of K-25 and K-27 onstream, it was decided to attempt raising the top to 60%. The plant was placed on total reflux with an elevated feed rate. To the surprise of practically no one, the assay rose to the target 60% in a short time. This level was maintained for approximately one year. During this time, the success of the recent past led to the decision to try for 93.15%, then being produced from the K-25 product by the electromagnetic process at the Y-12 Plant. In preparation for the assault, the amount of inventory required to sustain production at that level was calculated. Product at 60% was withdrawn and stockpiled for refeed. Upon reaching the necessary amount, the plant was again placed on total reflux accompanied by the refeeding of the 60% stockpiled material. The top assay reflected an increase but tended to level short of the target. It was determined that the cause for this was the buildup of U-234. Therefore, a small top withdrawal was initiated which resulted in reaching the U-235 target assay.

Daily material balances during the periods of change reflected substantial apparent losses particularly for U-235. It was recognized that the equipment surfaces were equilibrating to the higher enrichment exposure levels. Production requirements precluded any equipment offstream experimental activity until much later. Subsequent work utilizing the isotopic dilution technique (described later) revealed that the calculated inventory quantities in the separating equipment

were understated by as much as 20% of the reported value depending upon the location. The bases for inventory were not corrected but the experimental findings were used for qualifying material balance observations.

Several mathematical schemes of evaluating of the material loss were tried. Most notable among these was an equation of the form:

$$L = at + bT + c (F-W)$$

where: L = loss

a,b,c, are coefficients of:

t = time in days per balance interval

T = atmospheric temperature change per balance interval

(F-W) = feed minus waste production per balance interval

In retrospect, what was seen as having significant coefficients were:

t = long term loss rate to hidden inventory

(F-W) = changes with respect to time really reflected power or pressure level changes which interpreted the effect of physical adsorption

T = while the coefficient was of little significance, it probably reflected a change in process gas temperature with ambient.

As new plants were constructed, calculated cell total volumes were verified by nitrogen calibration tests. While this technique is not very sophisticated, it is time consuming and must recognize the compressibility factor of nitrogen. In the 1950's, it became possible to remove equipment temporarily from service for isotopic dilution testing of the calculated inventory retention. This procedure utilizes the material balance as the basis of evaluation. The equation is in the form:

$$I_{x_1} + C_{x_c} = (I + C)_{x_2}$$

where I = unknown weight of gas, chemisorption, and
physical adsorption at assay x_1

C = weight of diluting charge at assay x_c

x_2 = equilibrium assay

The above equation reduces to:

$$I = C \left\{ \frac{x_2 - x_c}{x_1 - x_2} \right\}$$

Two dilution tests are run. The first evaluates the term referred to as I. After evacuating and purging the system, a second test determines the amount of chemisorption. Physical adsorption cannot be independently determined but must be calculated for each test condition.

In order to run the isotopic dilution tests, it is desirable to have inverse recycle lines. These destroy the isotopic gradient which permits the determination of an equilibrium assay and prevent the accumulation of "lights" at the top of the cell. All K-25 and K-27 equipment included both 'A' and 'B' inverse recycle lines. However, these were not provided in the axial equipment buildings. One cell in the K-29 building was modified by installing one line for a test. The isotopic gradient was not completely flattened. Therefore, the equilibrium assay was determined by obtaining samples across the cell followed by graphical integration. Results of the test indicated the calculated inventory to be in error by only 0.19% when expressed as:

$$\frac{\text{Calculated inventory} - \text{Measured inventory}}{\text{Calculated inventory}} \times 100$$

Other tests in the K-25 and K-27 equipment are not enumerated herein since these buildings are no longer in service.

Isotopic dilution testing can be performed in some K-31 and K-33 cells where entire units are in standby. In these locations, the cell bypass lines can be utilized as inverse recycle lines.

WDMc:ht Reference K-12-735

2-18-71

Material Unaccounted For

We will now try to answer three questions:

CHART 1

1. What do we mean by "MUF" or material unaccounted for -- particularly in the context of Mr. Keller's letter -- how are these figures obtained, what do they include, and what are some of the variables associated with them?
2. What has been our recent MUF history, particularly since K-25 and K-27 were shut down in August of 1964.
3. What action can we take to:
 - a. Dampen the month to month variations in MUF, and
 - b. Insure that we safeguard our inventory over the long run?

What is MUF?

CHART 2

In simplest terms, MUF is the monthly material balance of the cascade -- the difference between the beginning and ending inventories adjusted for the inputs of feeds and withdrawals of product and tails from the cascade.

MUF is calculated both on the basis of kgs of uranium and kgs of U-235.

Of interest, the determination of separative work is dependent upon several of the same quantities used in the MUF calculations; thus variations in MUF will be reflected in variations in our separative work figure. This relationship is discussed in some detail in ORO-664, the report of the three-plant separative work study committee which met in 1968.

CHART 3

Looking first at the flows in and out of the cascade, the two primary feeds are Paducah product -- now 30 to 40 cylinders per month, and normal or toll material which may be from 30 to 50 cylinders per month, depending upon the mix of 2 1/2-10- and 14- ton containers.

Output from the cascade is either product to toll customers or to stockpile, about 18 cylinders, and tails to Paducah, roughly 50 to 60 cylinders per month. Thus, the typical month involves about 150 transfers in and out of cylinders.

1. WHAT IS "MUF"--MATERIAL UNACCOUNTED FOR?

A. HOW OBTAINED ?

B. WHAT DOES IT INCLUDE ?

C. VARIABLES ?

2. WHAT HAS BEEN OUR "MUF" HISTORY IN RECENT YEARS (SINCE K-25 AND K-27 SHUTDOWN IN AUGUST '64)

3. WHAT ACTION IS--AND CAN BE TAKEN--TO

A. DAMPEN MONTHLY VARIATIONS IN "MUF"

B. SAFEGUARD INVENTORY

MATERIAL BALANCE EQUATIONS

$$MUF_U = BI_U + F_U - EI_U - P_U - T_U$$

$$MUF_X = BI_X + F_X - EI_X - P_X - T_X$$

$$\text{SEPARATIVE WORK} = BI \cdot V(x) + F \cdot V(x) - EI \cdot V(x) - P \cdot V(x) - T \cdot V(x)$$

WHERE:

MUF = MATERIAL UNACCOUNTED FOR

BI = BEGINNING INVENTORY

EI = ENDING INVENTORY

F = FEED

P = PRODUCT

T = TAILS

$$V(x) = (2x-1) \ln(x/(1-x))$$

WITH SUBSCRIPT

U = URANIUM

X = URANIUM-235

CASCADE PERIMETER FLOWS

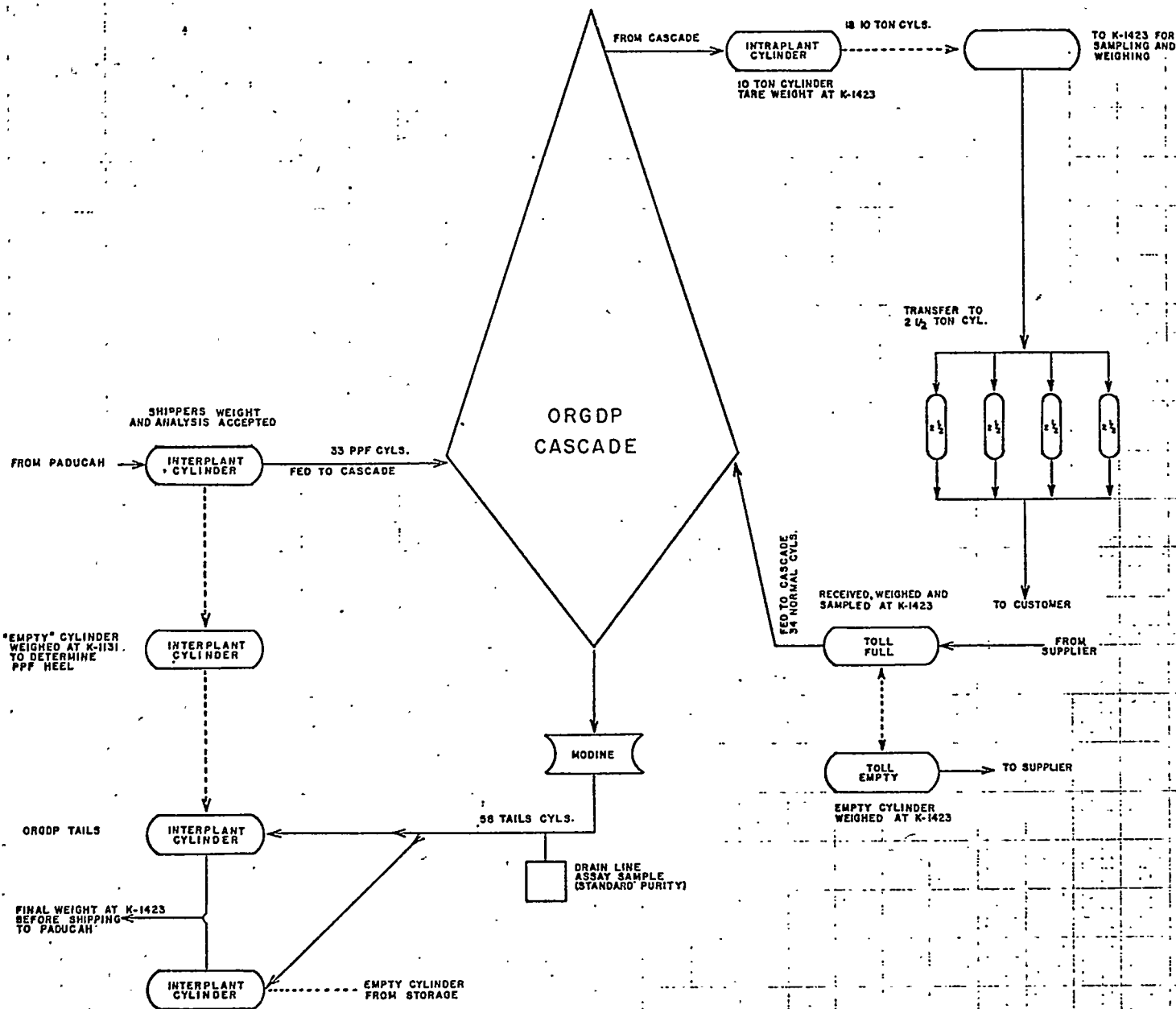


CHART 3

For simplicity, we've omitted side feed and withdrawals which parallel the above flows.

How good -- how accurate -- is our knowledge of these flows to and from the cascade? The answer varies with the particular stream you are considering.

In the case of Paducah Product, we accept, without question, their net weight, tare weight, analysis, and assay. We have recently, however, started a limited random check to see how our scales and lab results compare with theirs; for instance on the four cylinders checked in February, Paducah's gross weights were consistently above ours by an average of four pounds per cylinder. This bias, however, represents a difference of less than .02 of 1% and therefore, we have not increased our sampling plan. We have also found that Paducah tare weights appear to be somewhat overstated which in turn effects our heel calculations and results in a loss of our stated U-235 inventory of approximately 1/2 kg per month or somewhat less than 10% of our historical monthly U-235 MUF.

Our tails flow back to Paducah, usually in the same cylinders Paducah sends us, includes a gross check on the assay through a drain line sample and determination of net weight again based on historical tare weights.

The product withdrawal, on the other hand, receives considerable more attention. Here samples are taken from each cylinder, and weighing is done on K-25's best available scale. The same care is taken with toll material coming in from customers.

Looking at the perimeter activities as a whole, there are six areas with potential impact upon MUF:

1. Scale accuracy and bias (both here and at Paducah).
2. Sampling -- whether a sample is representative of the cylinder from which it is taken.
3. Cylinder tare weights and our subsequent assumptions regarding heel materials within the cylinder.
4. Laboratory analyses for assay and impurities.
5. Physical problem of instantaneously stopping material flows at the time of inventory.
6. Errors in gathering, transcribing, keypunching, and manipulating over 600 pieces of data each month that describe these parameter activities.

CHART 4

Now, looking at the cascade itself, not only do we have a flow of gaseous UF_6 in inventory (shown as the bottom area on the cross section) but also we have:

1. A film of UF_6 physically absorbed on equipment walls which is directly proportional to both surface area and pressure and inversely proportional to temperature.
2. A varying amount of UF_5 which goes back to UF_6 by fluorine atom exchange, a process we call active chem absorption,
3. Deposits of UO_2F_2 due to hydrolysis which we call inactive consumption.

The film of UF_6 physically absorbed on equipment walls is relatively small, approximately 600 kgs. This is released as gaseous UF_6 when the equipment is evacuated.

Likewise, the quantity of UF_5 active chem absorption is also small, estimated at approximately 700 kgs. for the K-25 cascade. This includes some material on surfaces of stand-by equipment which was not decontaminated in place at the time it was de-activated.

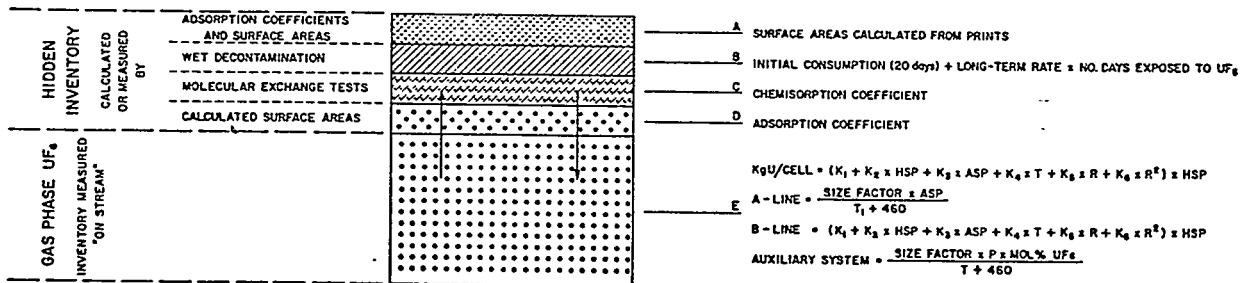
The deposits of UO_2F_2 inactive consumption, however, are considerable and again including material on the surfaces of stand-by equipment, amounts to over 12,300 kgs of U. This inactive consumption contributes both to monthly variations and long term losses since when equipment is taken out of service and decontaminated in K-1420 we are unable to go back and credit MUF with the material recovered. We will discuss its impact on MUF in more detail in a moment.

Calculations of our gas phase UF_6 are based upon 6600 pieces of data of which 5600 are directly associated with the basic cascade-- that is, they are readings of high-side and A-suction pressure, stage and B-outlet temperatures, lab assays, line recorder readings, and gas chromatograph results. The other 1,000 readings come from auxiliary systems.

This data is used in a series of equations which were developed in the early 1950's. The accuracy of these equations is also dependent upon:

1. The internal volumes of all pieces of equipment and associated piping, and
2. A series of coefficients based upon empirical tests and correlation studies.

INVENTORY METHODS



- A. EQUIPMENT WALL
- B. INACTIVE CONSUMPTION: UO_2F_2 , MATERIAL DEPOSITED ON EQUIPMENT WALL AS A RESULT OF HYDROLYSIS OF UF_6 .
- C. ACTIVE CHEMISORPTION: UF_6 , MATERIAL DEPOSITED ON EQUIPMENT WALLS WHICH WILL RETURN TO THE UF_6 GAS PHASE BY CAPTURING FLUORINE ATOM EXCHANGE FROM THE UF_6 GAS.
- D. PHYSICAL ADSORPTION: UF_6 , FILM OF MATERIAL ON EQUIPMENT WALL. WILL NOT EXIST IN THE ABSENCE OF GAS PHASE INVENTORY.
- E. GAS PHASE INVENTORY: UF_6

In a typical inventory, operators take instrument readings on the last shift of each month beginning roughly at 8:00 p.m. and ending at midnight. The data sheets are submitted to CTC keypunch at about 1:00 a.m. and the resultant cards are run through the barrier plant IBM 1800 by 10:00 a.m. the same morning.

Besides the obvious potential for incorrect readings, transcribing, keypunching other areas which can introduce an erroneous MUF are:

1. Changes in equipment or piping configurations which affect volumes.
2. Possible inventory equation errors or biases.
3. Cascade transients (the cascade is not stable while inventory readings are being taken).
4. Unknown releases or condensations since the last inventory.

With this brief background, let's look at our MUF experience over the past 6 1/2 years since the plant shut down K-25 and K-27.

CHART 5

We have put through 41,257,755 kgs of uranium and accounted for 41,236,892 kgs. The difference, 20,863, represents .05 of 1%.

Likewise, we have put through 250,382 kgs of U-235 and have accounted for 249,876 kgs. The difference 506 kgs represents .2 of 1%.

As mentioned earlier, MUF is somewhat of a misnomer since indeed we can account for some of the quantities reported as MUF. For instance:

1. Decontamination recovery
2. Hidden inventory
3. Known releases
4. Losses to the environment.

While these figures are not really as precise as the number of significant figures might indicate, it is probable that somewhere between 40 and 60% of MUF can be accounted for from these four sources of losses.

Our true MUF then would be on the order of .03 of 1% for uranium and .12 of 1% for U-235.

CASCADE DEFICIENCY SINCE AUGUST '64

	<u>MATERIAL HANDLED</u>	<u>REPORTED MUF</u>
U	41,257,755 KGS	20,863 KGS 0.05% (270 KGS/MO.)
U 235	250,382 KGS	506 KGS 0.2% (6.6 KGS/MO.)

KNOWN LOSSES (KGS)	<u>U</u>	<u>U235</u>
DECONTAMINATION RECOVERY	2242	75
INCREASED HIDDEN INV.	4676	49
REPORTED RELEASES	89	—
LOSS TO ENVIRONMENT *	2442	59
	<u>9449</u>	<u>183</u>

BALANCE OF MUF	11,414	323
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% ACCOUNTED FOR INCLUDING ABOVE KNOWN LOSSES	99.97%	99.88%
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GROSS % ACCOUNTED FOR FY T1 TO DATE	99.96%	99.83%
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* EST. OF ^{325 KGS}~~716 LBS~~/YR TO WATER
~~123 LBS~~/YR TO ATMOSPHERE
 42 KGS

Our gross or uncorrected MUF so far this fiscal year, that is, the last eight months has amounted to 1,888 kgs of U and 76 kgs of U-235. Our through put has been 4,229,339 kgs and 43,897 kgs of U and U-235 respectively, which results in a MUF loss of .04 of 1% for U and .17 of 1% for X. Another way to express this is that we have been able to account for 99.96% of U and 99.83% of U-235. These figures, however, do not include any adjustments for decontamination recovery, hidden inventory, reported releases, or losses to the environment. If we were to include these known losses, our MUF experience in the last eight months would be roughly 20% better than the 6 1/2 year average. It's dangerous, however, to draw any grand conclusions for such a relatively short period.

In terms of dollars, our unexplained loss experience since August, 1964, could range between 1.05 and 2.82% U-235. Its associated value ranges between \$7,000 and \$30,000 per month or over the last six years, our unexplained MUF totals between 1/2 and 2.3 million dollars.

We have been talking about averages over several years' time -- what about the monthly figures reported to the AEC.

CHART 6

This is simply a monthly plot of reported MUF. The vertical red lines show the changes in power levels from 1150 Mw to 1060 to 1080 to 500. We've also identified known causes for MUF such as condensations, incorrect datum pressures and unmeasured tails containers.

In addition to the possible sources of error already mentioned, other variations in monthly inventories are introduced by:

1. Inability to accurately account for test loop inventory unless the test loop is completely evacuated.
2. Variations within the purge cascade system - the amount of light gases determining the position of the UF₆ inventory.
3. Unknown quantities and assays of uranium in alumina and sodium chloride traps.
4. The varying effects of hidden inventory -- for example, when we increase pressure, we tend to increase MUF then gain it back when decreasing pressure. While the inverse is true with regard to temperature. Hidden inventory is not excluded from the reported MUF, however, since we can only measure it indirectly and cannot accurately account for such operations as unplugging of in-place barrier.

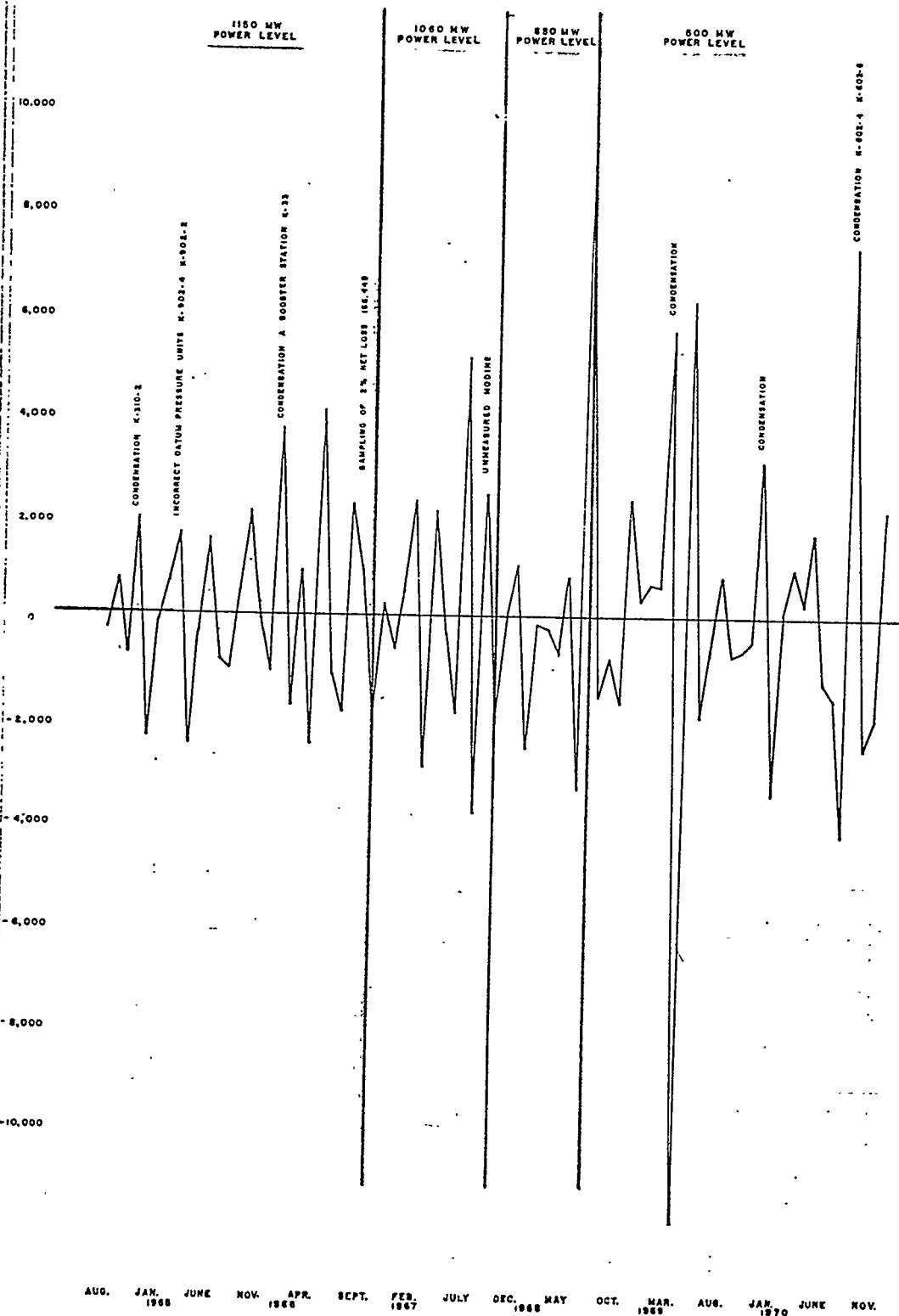


CHART 6

One of the things we are doing now is trying to measure the potential effect of each of these operations upon MUF.

CHART 7

A similar random pattern is found for the monthly MUF of U-235.

CHART 8

The black or top line shows the cumulative MUF over the last 6 1/2 years. As mentioned earlier, this totals 20,800 kgs. The green line is simply a least squares fit of the data for each of the four power levels we have had.

While we were at the 1150-1060 Mw level, a period of 38 months, the losses appeared to average about 400 kgs per month.

During the succeeding nine months, when the plant was at 880 Mw, we actually showed an average gain back of 350 kgs of U per month. This gain in inventory, however, was eliminated when we went to 500 Mw and for the last 29 months we have had a loss rate of about 120 kgs per month.

While we have not been able to identify a specific cause for the gain of U from October, 1967, to January, 1968, it is likely that there was a gross error in the handling of the data during this time.

The red line represents MUF if we account for known measured wet decontamination and the brown, or lower line, is an estimate of hidden inventory.

CHART 9

If we subtract out the four known major contributors to MUF -- the material recovered from decontamination, hidden inventory, known releases, and releases to the environment -- and use quarterly average figures to smooth out, somewhat, month to month variations, we get a loss rate averaging about 150 kgs U per month over the past 6 1/2 years.

CHART 10

The variations in U-235 show a very similar pattern -- a loss of 7 kgs during the first period after shutting down K-25 and K-27, followed by a leveling off and then a gaining back of 3 kgs per month and, during the last 2 1/4 years, an average loss rate of 5 1/2 kgs.

MONTHLY MUF
Kg U-235

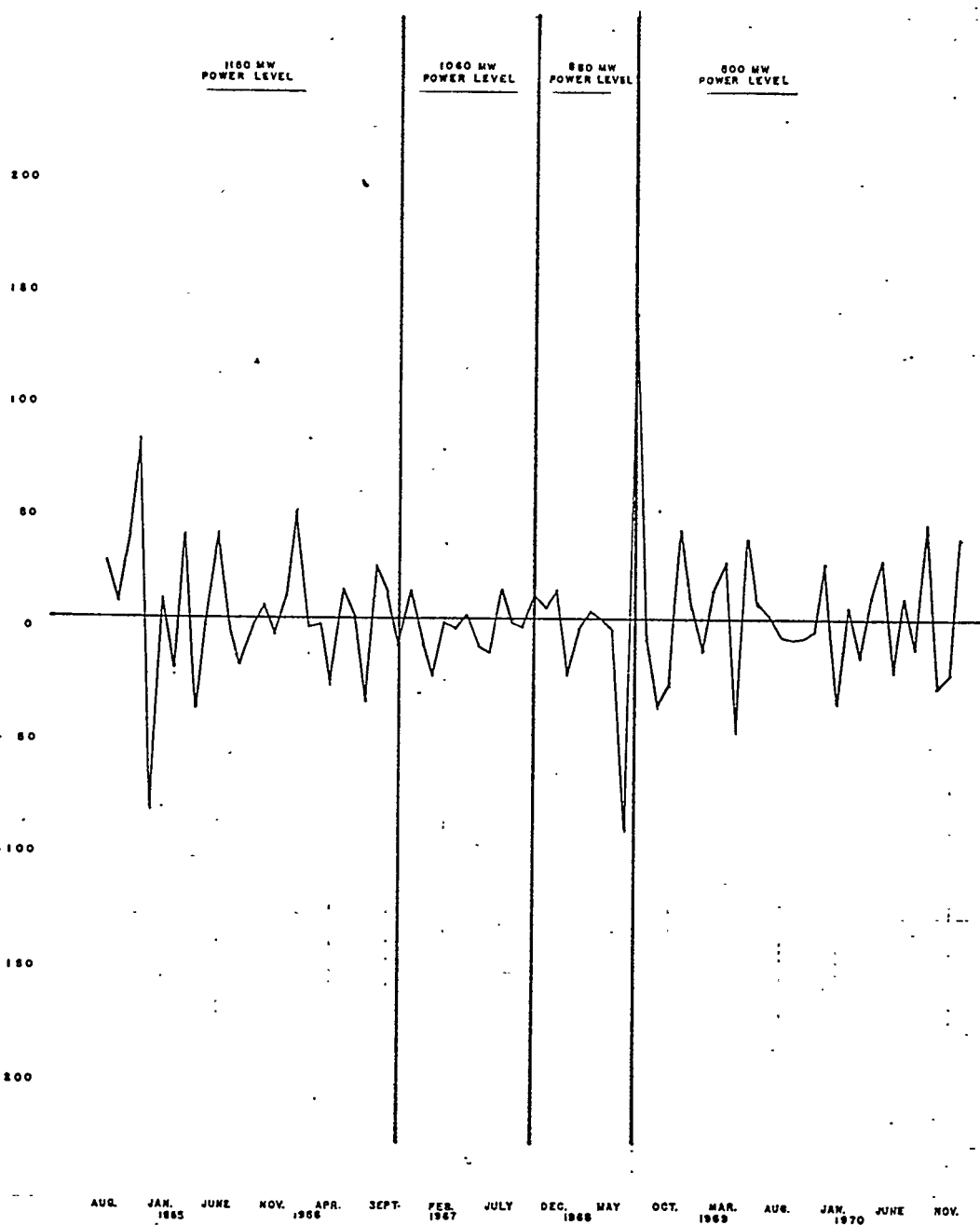


CHART 7

CUMULATIVE MUF
Kg U

ADJUSTED

HIDDEN INVENTORY QUANTITIES

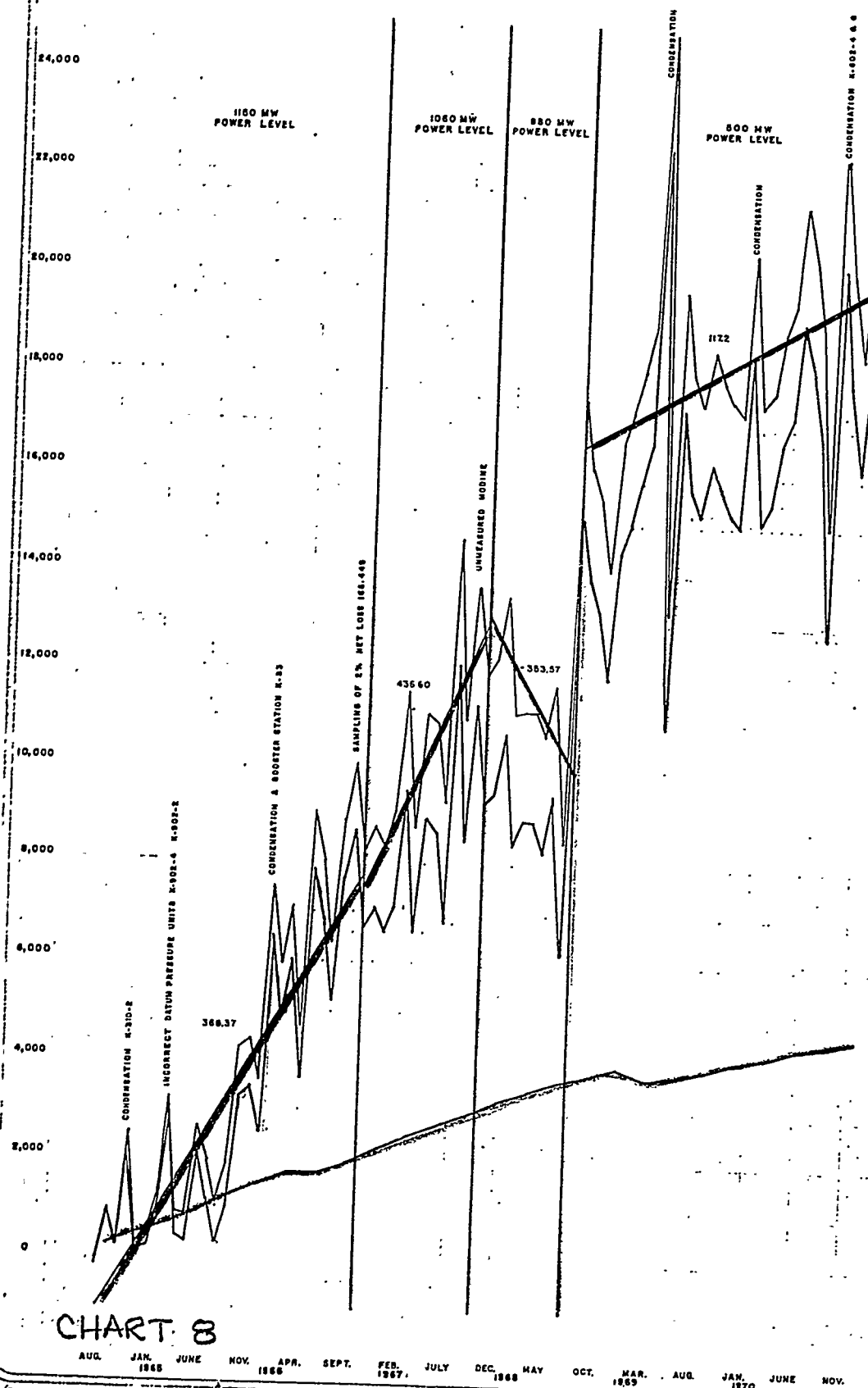


CHART 8

ADJUSTED CUMULATIVE MUF (U)

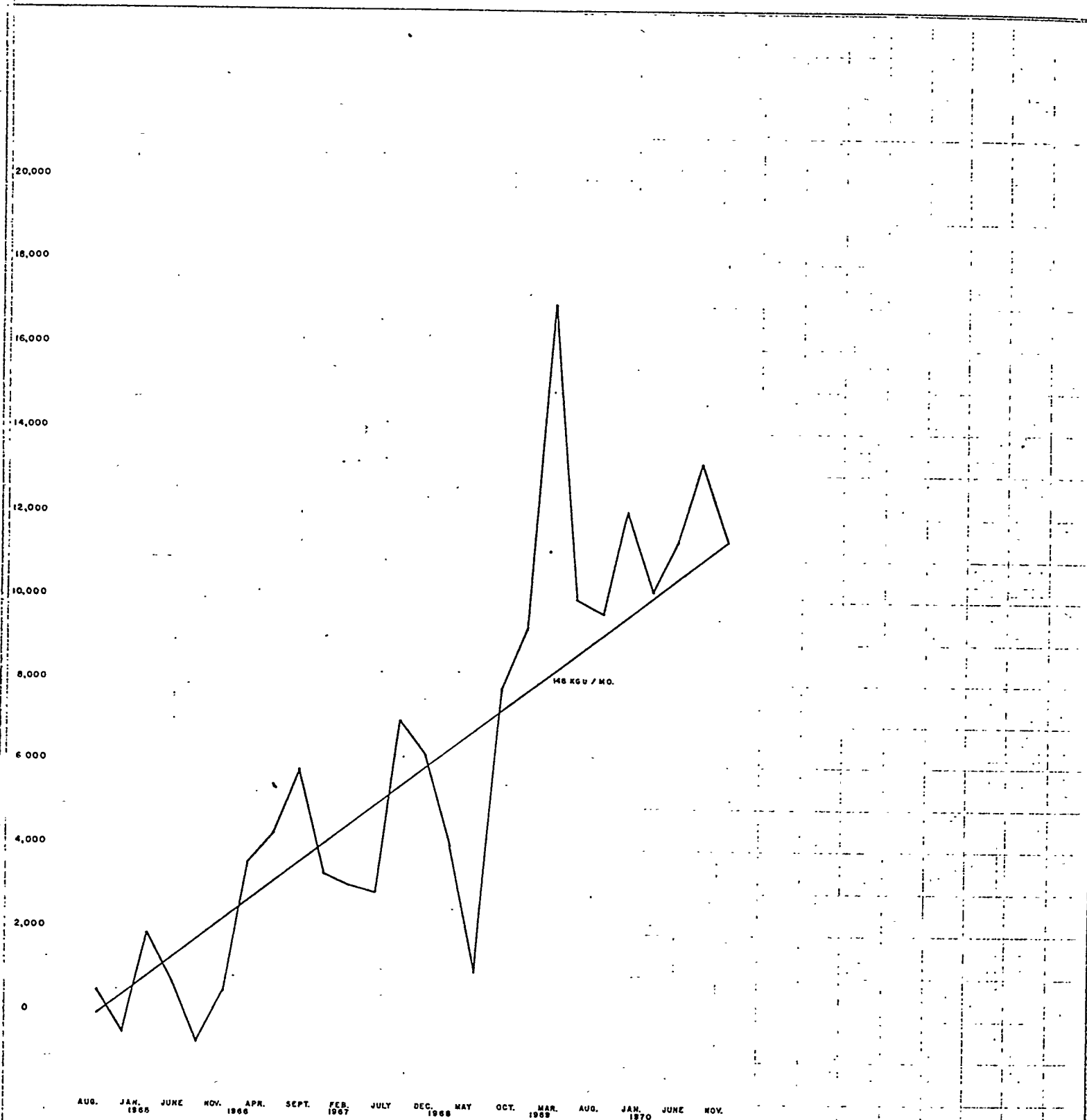


CHART 9

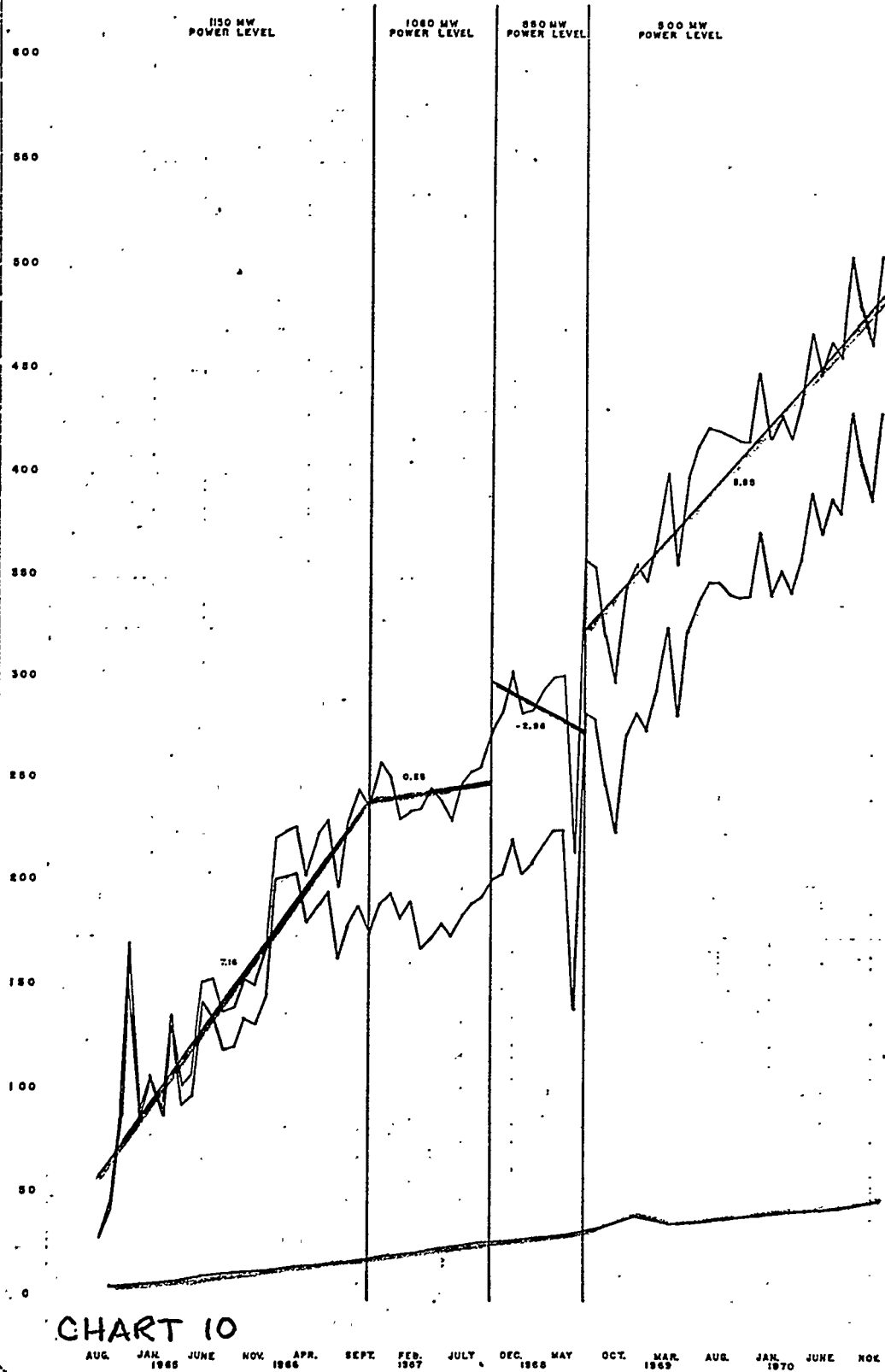


CHART 10

AUG. 1965 JAN. 1966 JUNE 1966 NOV. 1966 APR. 1967 SEPT. 1967 FEB. 1968 JULY 1968 DEC. 1968 MAY 1969 OCT. 1969 MAR. 1970 AUG. 1970 JAN. 1971 JUNE 1971 NOV. 1971

We started out saying that we would try to answer three questions.

1. What is MUF?
2. What has been our recent history?
3. What action can we take to both dampen our month to month variations in MUF and insure that we can reasonably account for our cascade inventory over a long period of time.

Addressing ourselves to the last question, let's go back and review the sources of loss and variation in MUF.

CHART 11

Any or all of the following may, to varying degrees, affect MUF:

1. Errors in handling some 7200 different pieces of data each month -- the potential effect upon MUF here can be major.
2. Releases and condensations -- can be major.
3. Test loop activities -- unknown effect, probably minor.
4. Purge cascade system -- unknown, probably moderate to minor.
5. Cascade transients -- unknown, probably minor.
6. Scale accuracy and bias at K-25 and Paducah -- moderate to minor.
7. Sampling -- that is, whether a sample is representative of the cylinder from which it is being taken and whether the assumptions on gradient within the cascade are correct -- moderate to minor.
8. Incorrect cylinder tare weight information -- minor effect.
9. Incorrect internal volumes or changes in equipment or piping configurations which, in turn, affect volumes -- unknown, potentially great but probably minor.
10. Inventory equation errors and biases -- probably minor.
11. Variations in flow cut-off times while taking inventory -- unknown, probably moderate to major effect.
12. Effects of hidden inventories (UF_5 , UO_2F_2 , and absorbed UF_6) -- unknown, likely moderate.
13. Accuracy in bias of assay and impurity analysis, and accuracy and bias of instrumentation -- unknown effect, probably minor.
14. Incomplete inventory information (for example, assumptions regarding such things as changes in temperatures and pressures within piping) -- probably moderate to minor depending upon random canceling effects.

SOURCES OF MONTHLY MUF VARIATIONS

1. HANDLING 7,200 PIECES OF DATA/MONTH
2. RELEASES AND CONDENSATIONS
3. TEST LOOP ACTIVITIES
4. CASCADE PURGE SYSTEM
5. CASCADE TRANSIENTS
6. WEIGHING SCALES -- ACCURACY AND BIAS
7. SAMPLING
8. CYLINDER TARE WEIGHTS
9. CASCADE VOLUMES
10. INVENTORY EQUATIONS
11. FLOW CUT-OFFS
12. HIDDEN INVENTORIES
13. ANALYSIS AND OTHER MEASUREMENTS
14. INCOMPLETE INFORMATION

CHART 11

What can be done to minimize these variations:

1. Increased awareness, planning, and follow-through on taking inventory and handling the associated data. Here, we are:
 - a. simplifying the forms upon which data will be taken.
 - b. making dry runs before taking the monthly inventory and holding critiques within five days afterward.
 - c. minimizing the number of hand calculations by increasing our use of computer facilities.
 - d. utilizing the cascade coordinator to supervise taking of inventory data.
 - e. establishing checks and diagnostic routines to highlight month to month inconsistencies and identify potential errors (Paducah has already done considerable work in this area).
 - f. we took a triple inventory at the end of February to analyze variations in shift to shift activities.
2. We are taking advantage of the results of Bob Jordan's committee on environmental studies to better quantify our routine and extra-ordinary losses to the surroundings.
3. We have developed a program to calculate a weekly inventory which, while not as accurate in an absolute sense as the end-of-the-month inventory, will show relative changes, thereby identifying problems and allowing sufficient time to take corrective action prior to the end-of-the-month inventory (which must be submitted to the AEC within five working days thereafter). We are also maintaining a daily graph of the difference between input and output to and from the cascade which has proven valuable in identifying condensations and other major deviations.
4. For the February inventory, we closed down and evacuated the test loop (although we do not plan to make this a routine procedure each month).
5. A task force is already working on modifications to the purge cascade and as their work progresses, the impact upon inventory accuracy will also be reviewed.
6. Data was taken during the last inventory to determine the potential effect of transients -- further analysis is planned.

7. We have started a modest random sampling plan to compare Paducah's weights and analyses with our values.
8. We have done extensive analysis on the variance of our own scales, particularly in K-1423, the toll enrichment area. Work done by Y-12's Pat Reavis indicates error less than ± 1 lb. in 20,000 pounds and less than 2 pounds in 40,000 pounds.
9. A statistical plan has been developed for determining variance in sampling and we are proceeding on a small scale to better understand the magnitude of this problem and its associated cost.
10. While correcting tare weights on cylinders is pretty much a function of the dollars available, we are maintaining a record of the effect that these incorrect tare weights have upon MUF (for example, in February 1/2 kg U-235).
11. Regarding internal volume and surface area calculations, several man-years of effort are being devoted to a complete overhaul of this data, both updating present prints and providing a basis for rapid and accurate calculation of new volumes and surface areas for CIP. Ray Greene of Don Kellogg's group in Engineering is doing many of these calculations on a time-shared computer system, developing routines for T's, junctions, double-sweep T's, etc. As an indication of the size of this project, 1500 drawings are being reviewed and evaluated to complete the basic piping sub-assemblies for K-33.
12. Although we do not have immediate plans for extensive work on revising inventory equations, we do recognize that we have had only one verification of these equations for axial equipment, and that was made over ten years ago. Our approach here is to evaluate the cost and potential advantages of going into a more thorough review and are considering perhaps one assay dilution and molecular exchange test early next fiscal year.
13. With regard to cut-off times of material flows in and out of the cascade, it appears that considerable improvement can be made through better procedures and perhaps some additional personnel on an as required basis during the actual taking of the inventory.

14. Our approach to the evaluation of the effects of hidden inventories, UF_5 , absorbed UF_6 , and UO_2F_2 , again is to explore the costs and potential of doing more basic research in this area. With current accounting efficiencies between 99.8 and 99.9% we do not expect any great surprises here.
15. We feel that a modest number of experiments, carefully designed, will give us considerably more insight into the contribution of MUF variations due to analytical and instrument errors.
16. Operating groups and Operations Planning Department are on a selective basis resolving problem areas regarding cascade information being used in inventory calculations. This is a continuing effort by two to three people in conjunction with their other responsibilities.
17. We are planning to use the MONAL trailer to confirm hidden inventory assumptions in calculations when it arrives here next month. Initial plans are being developed now.

With regard to long term accountability, plans are also being made to evaluate the effect of CIP activities on MUF and develop procedures so that accurate information can be obtained regarding the material that is to be recovered through decontamination. We are also doing considerable statistical analysis in order to better understand the extent to which various areas have contributed to MUF and insure through daily and weekly monitoring that any gross deviations from past history are immediately recognized and can be effectively evaluated. Our philosophy is that careful daily attention to MUF will insure long term safeguarding of material.

A great deal of money could be spent in additional measuring devices, computers, and other hardware, as well as developing a larger staff. Our approach, however, is one of better utilizing what we have and, before we spend additional money, assuring ourselves that there will, indeed, be a potentially significant payoff.

(This section to be completed by subcontractor requesting document)

J. Lamb / 1034A
Requestor Document Center (is requested to provide the following document)

Date of request ~~02/28/95~~ 6/28/96 Expected receipt of document 1/8/95

Document number _____ Date of document 1970's

Title and author (if document is unnumbered)

The MUF Story

(This section to be completed by Document Center)

Date request received 7/1/96

Date submitted to ADC 7/11/96

Date submitted to HSA Coordinator 7/1/96

(This section to be completed by HSA Coordinator)

Date submitted to CICO 7/11/96

Date received from CICO 7/16/96

Date submitted to ChemRisk/Shonka and DOE 8/1/96

(This section to be completed by ChemRisk/Shonka Research Associates, Inc.)

Date document received _____

Signature _____

THE MUF STORY

Bill McCluen has presented an excellent background of cascade material control activities during the early years of the plant start-up and through the 1950's when most of the basic work was done to identify and measure uranium losses.

We will now try to answer three questions:

Chart 1

1. What do we mean by "MUF" or material unaccounted for -- particularly in the context of Mr. Kellar's letter -- how are these figures obtained, what do they include, and what are some of the variables associated with them?
2. What has been our recent MUF history, particularly since K-25 and K-27 were shut down in August of 1964.
3. What action can we take to:
 - a. Dampen the month to month variations in MUF, and
 - b. Insure that we safeguard our inventory over the long run.

What is "MUF"?

Chart 2

In simplest terms, MUF is the monthly material balance of the cascade -- the difference between the beginning and ending inventories adjusted for the inputs of feeds and the withdrawals of product and tails.

MUF is calculated both on the basis of kgs of uranium and kgs of U-235.

Of interest, the determination of separative work is dependent upon several of the same quantities used in the MUF calculations; thus, variations in MUF will be reflected in variations in our separative work figure.

This document has been approved for release
to the public by: *[Signature]*

[Signature]
Technical Information Officer
Oak Ridge K-25 Site

[Signature]
Date

Prepared by Union Carbide Corporation-
Nuclear Division, operating contractor for the
U.S. Department of Energy under U.S.
Government Contract No. W-7405-eng-26.

1. WHAT IS "MUF"-- MATERIAL UNACCOUNTED FOR?
 - A. HOW OBTAINED ?
 - B. WHAT DOES IT INCLUDE ?
 - C. VARIABLES ?
2. WHAT HAS BEEN OUR "MUF" HISTORY IN RECENT YEARS (SINCE K-25 AND K-27 SHUTDOWN IN AUGUST '64)
3. WHAT ACTION IS--AND CAN BE TAKEN--TO
 - A. DAMPEN MONTHLY VARIATIONS IN "MUF"
 - B. SAFEGUARD INVENTORY

MATERIAL BALANCE EQUATIONS

$$MUF_U = BI_U + F_U - EI_U - P_U - T_U$$

$$MUF_X = BI_X + F_X - EI_X - P_X - T_X$$

$$\text{SEPARATIVE WORK} = BI \cdot V(x) + F \cdot V(x) - EI \cdot V(x) - P \cdot V(x) - T \cdot V(x)$$

WHERE:

MUF = MATERIAL UNACCOUNTED FOR

BI = BEGINNING INVENTORY

EI = ENDING INVENTORY

F = FEED

P = PRODUCT

T = TAILS

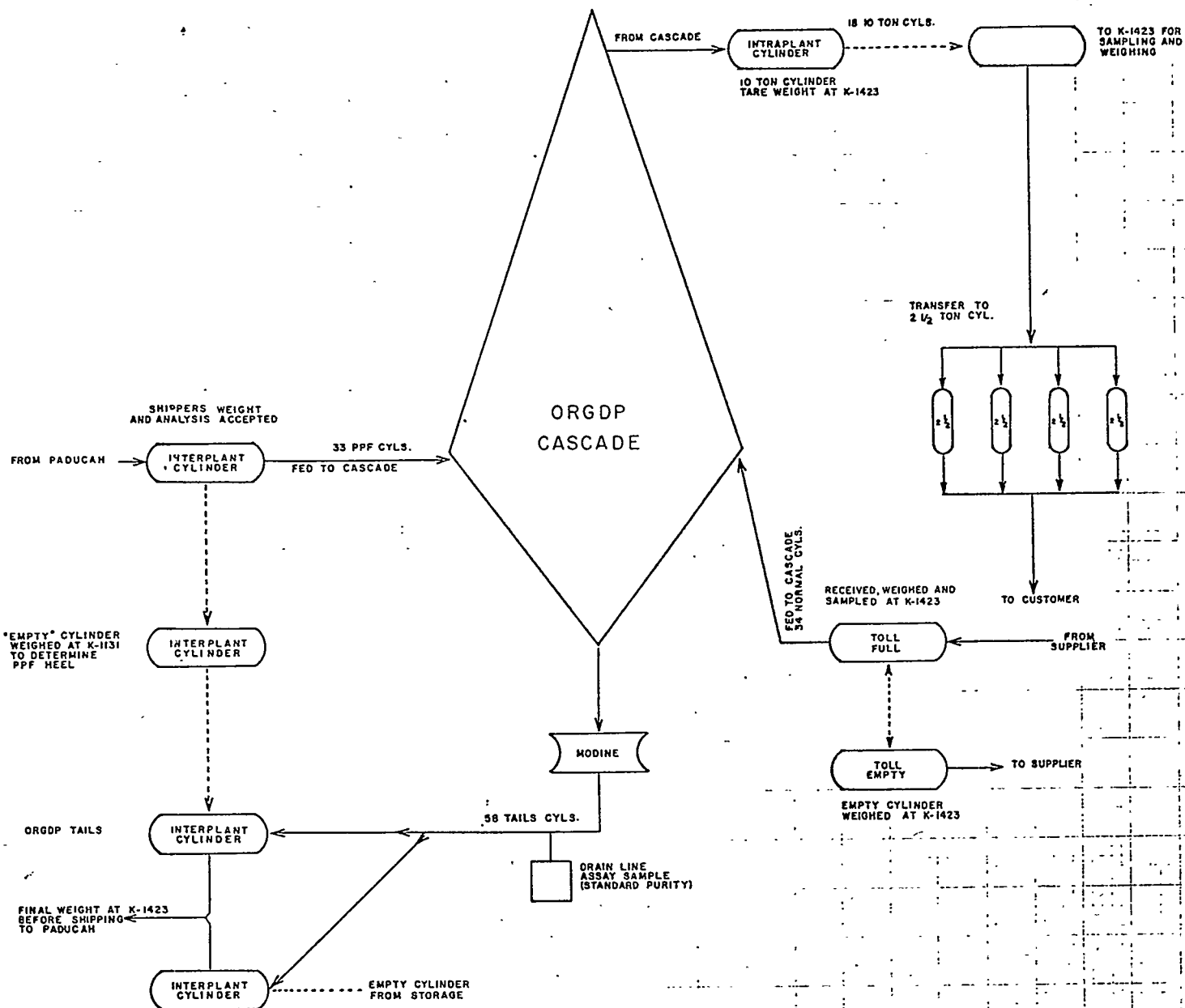
$$V(x) = (2x-1) \ln(x/(1-x))$$

WITH SUBSCRIPT

U = URANIUM

X = URANIUM-235

CASCADE PERIMETER FLOWS



Looking first at the flows in and out of the cascade, the two primary feeds are Paducah product -- now averaging 30 to 40 10- and 14-ton cylinders per month -- and normal or toll material -- which will be between 30 and 50 cylinders per month depending upon the mix of toll material which may be in 2 1/2 or 10- ton containers and normal material in 14-ton cylinders.

Output from the cascade is either product to toll customers or to stockpile-- presently about 18 10-ton cylinders per month -- and tails to Paducah -- roughly, 50 to 60 10- and 14-ton cylinders.

For simplicity, we've omitted side feed and side withdrawals which are very similar to the above operations.

How good -- how accurate -- is our knowledge of these flows to and from the cascade? The answer varies with particular stream you are looking at.

In the case of Paducah product, we accepted their net weight, their tare weight, their analyses and their assays. We have recently, however, started a limited random check to see how our scales and lab results compare with theirs. For instance, on the four cylinders checked in February, Paducah's gross weights were consistently above ours by an average of four pounds per cylinder. At this time, about the only thing we can say, however, is that their tare weights appeared to be somewhat overstated which in turn affects our heel calculation and results in a slight loss of our stated U-235 inventory.

Our tails flow back to Paducah generally in the same cylinders Paducah sends to us. Here we take a drain line assay sample which is a gross check and determine the net weight of product based again on their tare weights.

The product withdrawal, on the other hand, receives considerable more attention. Here samples are taken from each cylinder, and weighing is done on K-25's best available scale. The same care is taken with toll material coming in from customers.

Looking at the parameter activities as a whole, there are six areas with potential impact upon MUF:

1. Scale accuracy and bias (both here and at Paducah)
2. Sampling -- whether a sample is representative of the cylinder from which it is taken
3. Cylinder tare weights and our subsequent assumptions regarding heel materials within the cylinder
4. Laboratory analyses for assay and impurities
5. Physical problem of instantaneously stopping material flows at the time of inventory.
6. Errors in gathering, transcribing, keypunching, and manipulating over 600 pieces of data each month that describe these parameter activities.

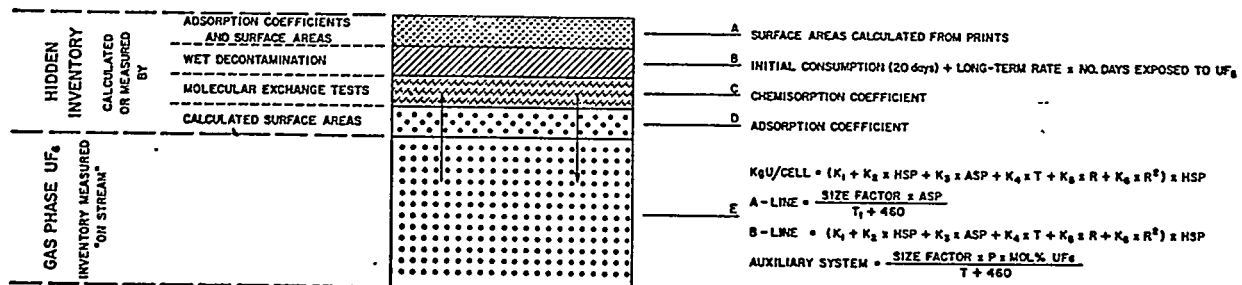
Chart 4

Now looking into the cascade itself, not only do we have a flow of gaseous UF_6 in inventory but also we have :

1. A film of UF_6 physically absorbed on equipment walls which is directly proportional to pressure and inversely proportional to temperature and which is released upon evacuation of equipment.
2. A varying amount of UF_5 which goes back to UF_6 by fluorine atom exchange (active chem absorption), and
3. Deposits of UO_2F_2 due to hydrolysis (inactive consumption) and which is recovered by in place or wet decontamination.

The UO_2F_2 contributes both to monthly variations and long term losses since, when equipment is taken out of service and decontaminated in K-1420 we are unable to go back and credit MUF with material recovered. We will discuss this in a little more detail in a moment.

INVENTORY METHODS



- A. EQUIPMENT WALL
- B. INACTIVE CONSUMPTION: UO_2F_2 , MATERIAL DEPOSITED ON EQUIPMENT WALL AS A RESULT OF HYDROLYSIS OF UF_6 .
- C. ACTIVE CHEMISORPTION: UF_6 , MATERIAL DEPOSITED ON EQUIPMENT WALLS WHICH WILL RETURN TO THE UF_6 GAS PHASE BY CAPTURING FLUORINE ATOM EXCHANGE FROM THE UF_6 GAS.
- D. PHYSICAL ADSORPTION: UF_6 , FILM OF MATERIAL ON EQUIPMENT WALL. WILL NOT EXIST IN THE ABSENCE OF GAS PHASE INVENTORY.
- E. GAS PHASE INVENTORY: UF_6

Calculations of our gas phase UF_6 is based upon 6600 pieces of data of which 5600 are directly associated with the basic cascade and include high side and A-suction pressure readings, stage and B-outlet temperatures, lab assays, line recorder readings, and gas chromatograph results. The other 1,000 readings come from auxiliary systems.

This information is used in a series of equations developed in the early 1950's to calculate inventory and, in addition to the monthly data gathered, are also dependent upon:

1. The internal volumes of all pieces of equipment and associated piping, and
2. A series of coefficients based upon imperical tests and correlation studies.

In a typical inventory, operators take instrument readings on the last shift of each month beginning roughly at 8:00 p.m. and ending at midnight. The data sheets are submitted to CTC keypunch at about 1:00 a.m. and the resultant cards are run through the barrier plant IBM 1800 usually by 10:00 a.m. the same morning.

Besides the obvious potential for incorrect readings, transcribing, and keypunching other areas having an impact upon MUF are:

1. Changes in equipment or piping configurations which affect volumes
2. Possible inventory equation errors or biases.
3. Cascade transients (the cascade is not stable while inventory readings are being taken)
4. Unknown releases or condensations since the last inventory

With this brief background, let's look at our MUF experience over the past 6 1/2 years since the plant shut down K-25 and K-27.

Chart 5

CASCADE DEFICIENCY SINCE AUGUST '64

	<u>MATERIAL HANDLED</u>	<u>REPORTED MUF</u>
U	41,257,755 KGS	20,863 KGS 0.05% (270 KGS/MO.)
U 235	250,382 KGS	506 KGS 0.2% (6.6 KGS/MO.)

KNOWN LOSSES (KGS)	<u>U</u>	<u>U 235</u>
DECONTAMINATION RECOVERY	2242	75
INCREASED HIDDEN INV.	4676	49
REPORTED RELEASES	89	—
LOSS TO ENVIRONMENT *	<u>2442</u>	<u>59</u>
	9449	323
BALANCE OF MUF	11,414	323
% ACCOUNTED FOR		
INCLUDING ABOVE KNOWN LOSSES	99.97%	99.88%

*EST. OF 716 LBS/YR TO WATER
123 LBS/YR TO ATMOSPHERE

We have put through 41,257,755 kgs of uranium and have accounted for 41,236,892 kgs. The difference 20,863 represents .05 of 1%.

Likewise, we have put through 250,382 kgs of U-235 and have accounted for 249,876 kgs. The difference 506 kgs represents .2 of 1%.

As alluded to earlier, MUF is somewhat of a misnomer since indeed we can account for some of the quantities reported as MUF. For instance:

1. Decontamination recovery
2. Hidden inventory
3. Known releases
4. Losses to the environment

While these figures are not really as precise as the number of significant figures might indicate, it is probably that somewhere between 40 and 60% of MUF can be accounted for from these four sources of losses.

Our true MUF then would be on the order of .03 of 1% for uranium and .12 of 1% for U-235.

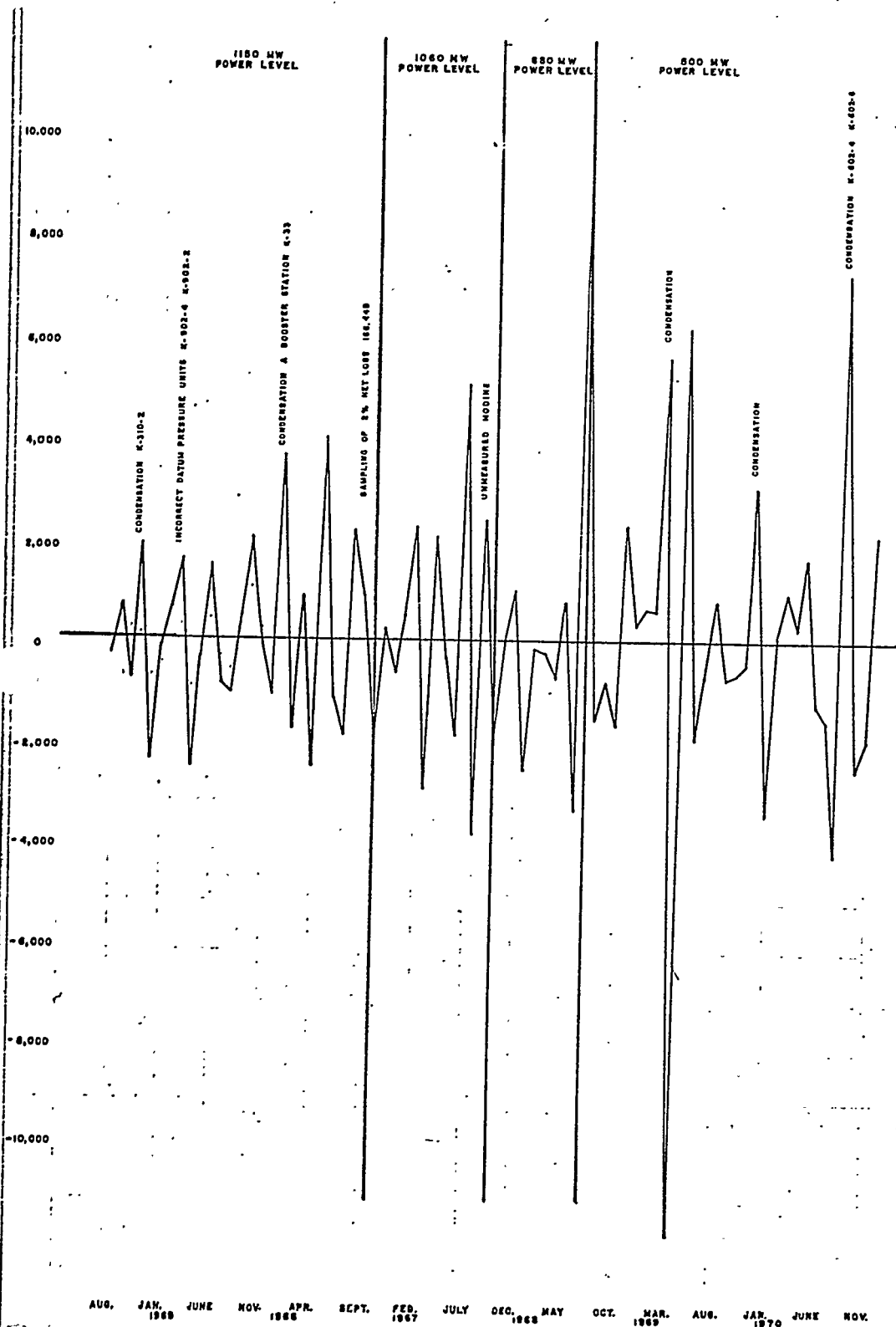
The average assay level of this unexplained loss could range between 1.05 and 2.82% U-235 and its associated value would have a comparable range between \$7,000 and \$30,000 per month or over the last six years our unexplained MUF would total between half a million and 2.3 million dollars.

We have been talking about averages over several years' time -- what about the monthly figures reported to the AEC.

Chart 6

This is simply a monthly plot of reported MUF. The vertical red line shows the changes in power levels from 1150 megawatts to 1060 to 880 to 500. We have also identified known causes for MUF such as condensation, incorrect datum pressures, and unmeasured tails containers.

MONTHLY MUF
Kg U



Other potential contributors to these variations in addition to those mentioned earlier are:

1. Inability to accurately account for test loop inventory unless the test loop is completely evacuated.
2. Variations within the cascade purge system -- the amount of light gases determining the position of the UF_6 inventory
3. Unknown quantities in assays of uranium in alumina and sodium fluoride traps.
4. The varying effects of hidden inventory -- for instance, when we increase pressure we tend to increase MUF and then gain it back when decreasing pressure which the inverse is true with regard to temperature.

One of the things we are trying to do now is to get a better handle on the potential effect of these various operations and phenomena.

Chart 7

A similar random pattern is found for the monthly MUF of U-235.

Chart 8

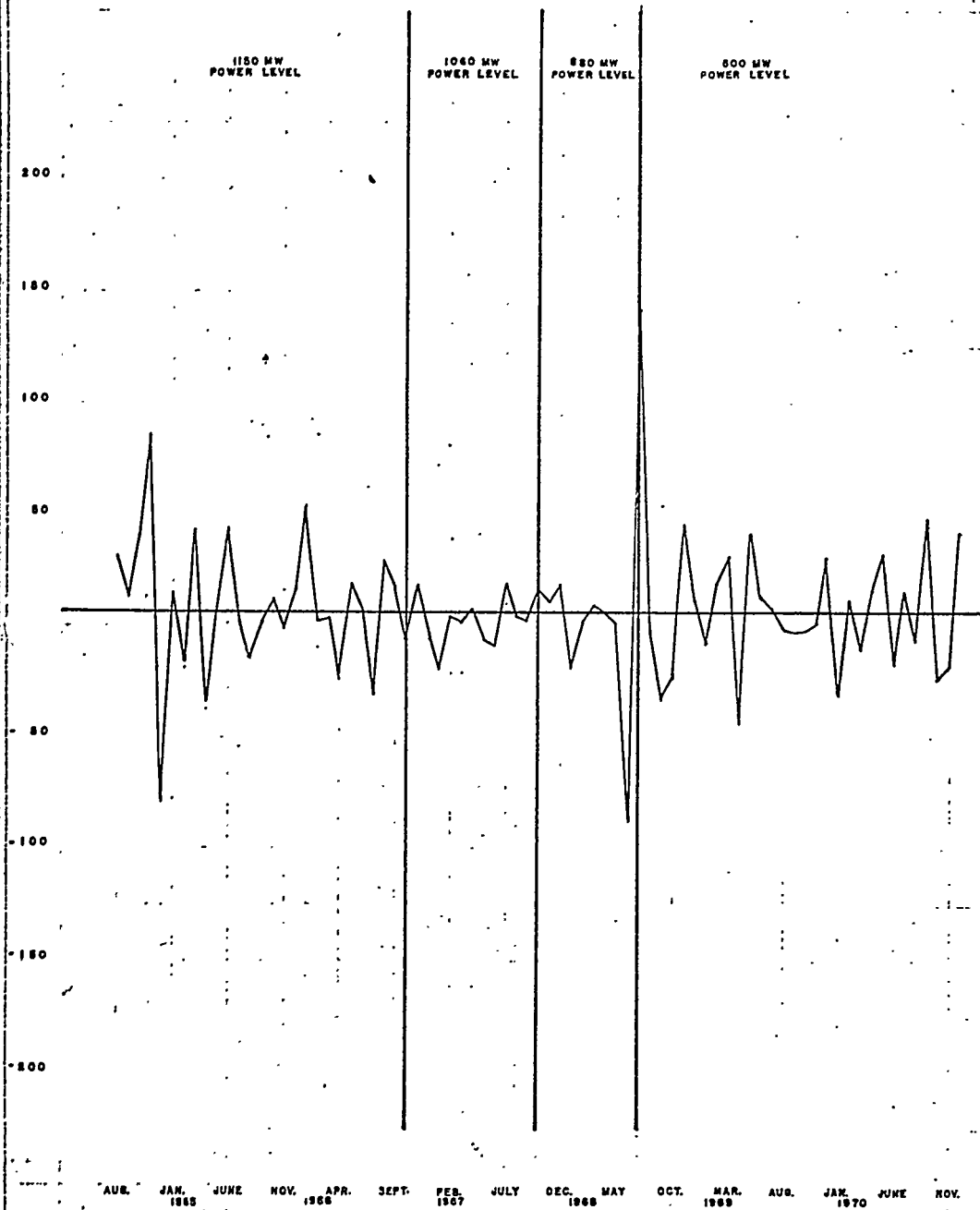
The black or top line shows the cumulative MUF over the last 6 1/2 years. As mentioned earlier, this totals 20,800 kgs. The green line is simply a least squares fit of the data for each of the four power levels we have had.

While we were at the 1150-1060 megawatt level, a period of 38 months, the losses appeared to average about 400 kgs per month.

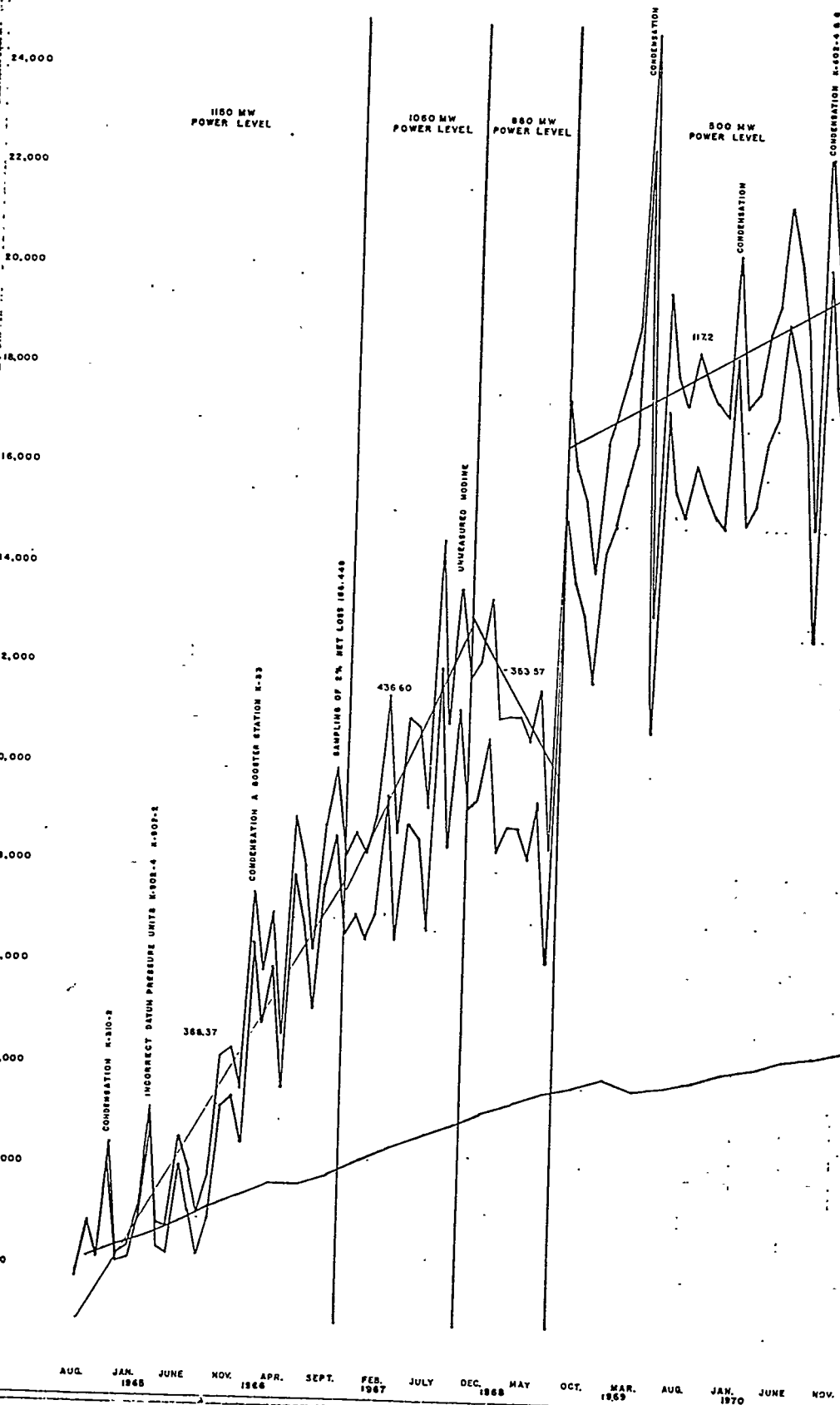
During the succeeding 9 months when the plant was at 880 megawatts we actually showed an average gain of 350 kgs per month. This gain in inventory, however, was eliminated when we went to 500 megawatts and for the last 29 months we have had a loss rate of about 120 kgs per month.

While we have not been able to identify a specific cause for the gain of U from October, '67, to January, '68, it is likely that there was a gross error in the handling of data during this time.

MONTHLY MUF
Kg U-235



CUMULATIVE MUF ——— ADJUSTED ——— HIDDEN INVENTORY QUANTITIES ———
Kg U



The red line represents MUF if we account for decontamination and the brown line or lower line is an estimate of hidden inventory.

Chart 9

If we subtract out the four known major contributors to MUF, the material recovered from decontamination, hidden inventory, known releases, and releases to the environment, and use quarterly average figures to smooth out somewhat month to month variations, we get a loss rate averaging about 150 kgs U per month.

Chart 10

The variations in U-235 show a very similar pattern -- a loss of 7 kgs during the first period after shutting down K-25 and K-27, followed by a leveling off and then a gaining back of 3 kgs per month and, during the last 2 1/4 years, an average loss rate of 5 1/2 kgs.

We started out saying that we would try to answer three questions.

1. What was MUF?
2. What has been our recent history?
3. What action can we take to both dampen out month to month variations in MUF and insure that we can reasonably account for our cascade inventory over a long period of time.

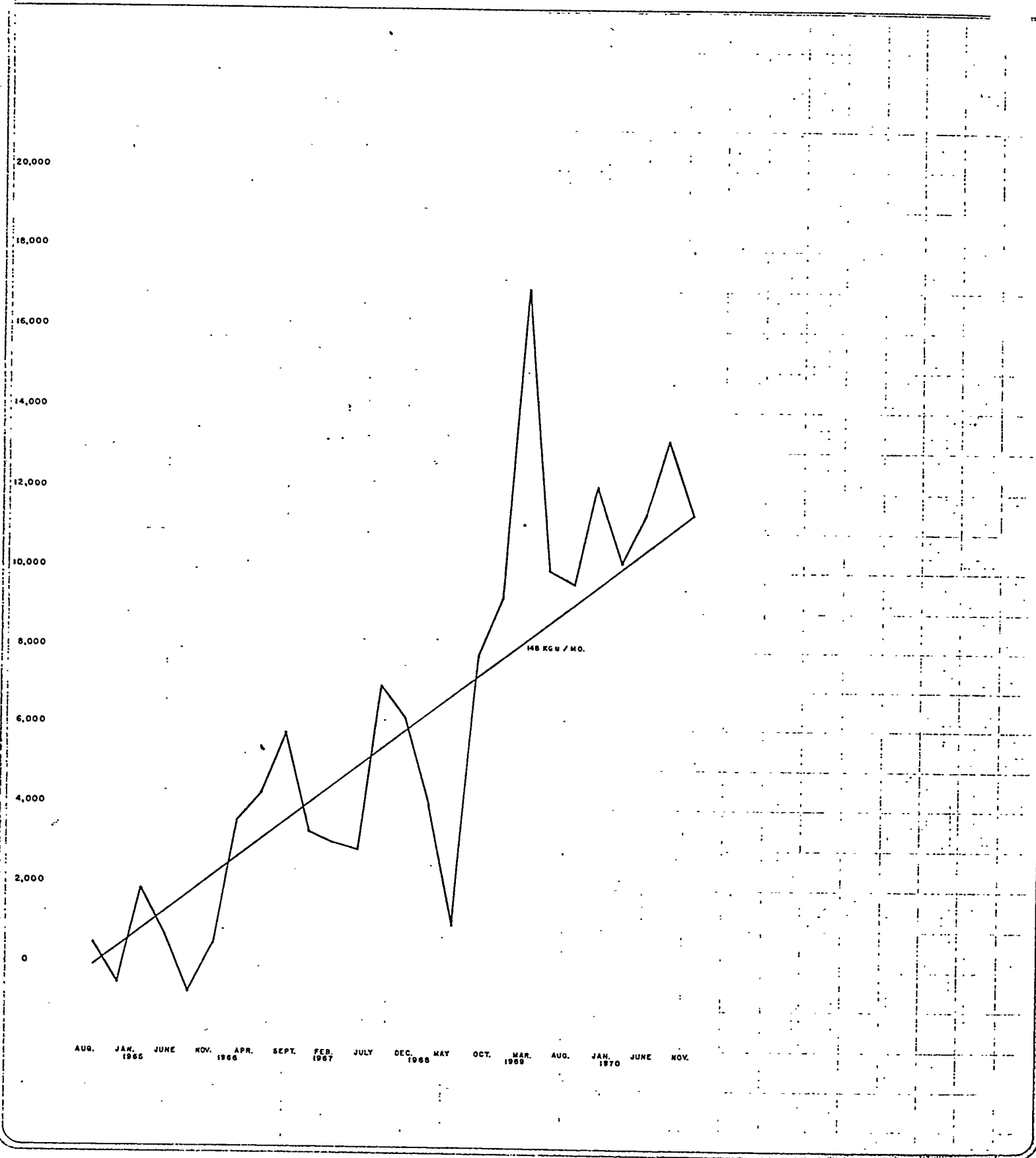
Let's go back and review the sources of losses and causes for variation in MUF.

Chart 11

Any or all of the following may, to varying degrees, affect MUF:

1. Errors in handling some 7200 different pieces of data each month
2. Releases and condensations
3. Test loop activities
4. Cascade purge system
5. Cascade transients

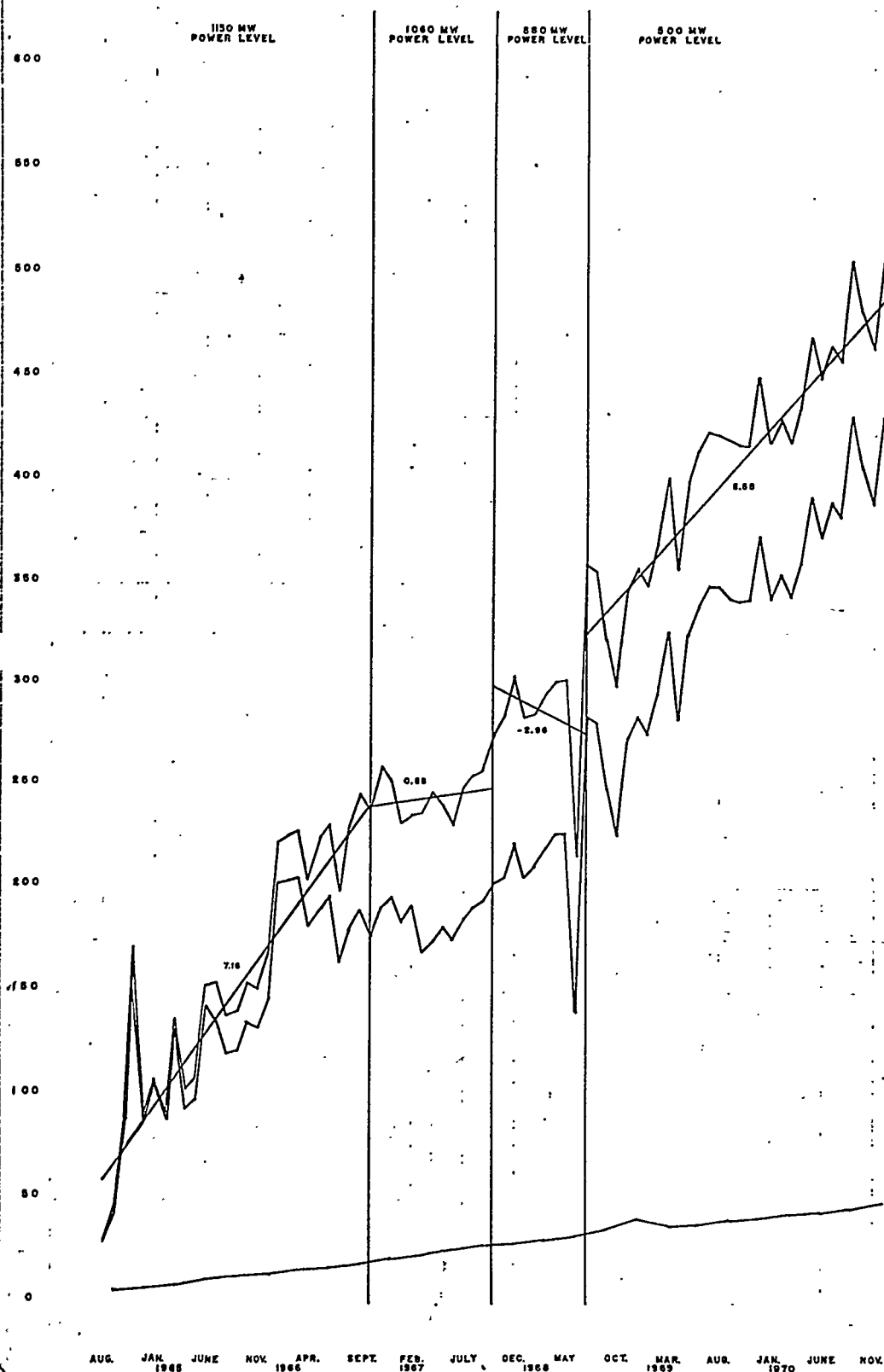
ADJUSTED CUMULATIVE MUF (U)



CUMULATIVE MUF
Kg U-235

ADJUSTED

HIDDEN INVENTORY QUANTITIES



SOURCES OF MONTHLY MUF VARIATIONS

1. HANDLING 7,200 PIECES OF DATA/MONTH
2. RELEASES AND CONDENSATIONS
3. TEST LOOP ACTIVITIES
4. CASCADE PURGE SYSTEM
5. CASCADE TRANSIENTS
6. WEIGHING SCALES -- ACCURACY AND BIAS
7. SAMPLING
8. CYLINDER TARE WEIGHTS
9. CASCADE VOLUMES
10. INVENTORY EQUATIONS
11. FLOW CUT-OFFS
12. HIDDEN INVENTORIES
13. ANALYSIS AND OTHER MEASUREMENTS
14. INCOMPLETE INFORMATION

6. The weighing of inputs and outputs -- scale accuracy and bias at K-25 and Paducah.
7. Sampling -- whether a sample is representative of the cylinder from which it is being taken and whether the assumptions on gradient within the cascade are correct.
8. Incorrect cylinder tare weight information.
9. Incorrect internal volumes or changes in equipment or piping configurations which, in turn, would affect volumes.
10. Inventory equation errors and biases.
11. Variations in flow cut-off times while taking inventory.
12. Effects of hidden inventories, UF_5 , UO_2F_2 , and absorbed UF_6 .
13. Accuracy and bias of assay and impurity analysis, as well as in other instruments.
14. Incomplete inventory information -- particularly, assumptions regarding such things as changes in temperatures and pressures within piping.

What can be done to minimize these variations:

1. Increased awareness, planning, and follow-through on taking inventory and handling the associated data. Here, we are:
 - a. Simplifying the forms upon which data will be taken.
 - b. Making dry runs before taking the inventory and holding critiques afterwards.
 - c. Minimizing the number of hand calculations by increasing our use of computer facilities.
 - d. Utilizing the cascade coordinator to supervise inventory taking.
 - e. We took a triple inventory at the end of February to analyze variations in shift-to-shift activities.
 - f. We are establishing checks and diagnostic routines to highlight month to month inconsistencies and identify potential errors.

2. We are taking advantage of the results from Bob Jordan's committee on environmental studies to better quantify our routine and extraordinary losses to the surroundings.
3. We are planning to take weekly "mini-inventories" to identify problems and have sufficient time to take corrective action prior to the end of the month inventory which must be submitted to the AEC withing 5 working days.
4. For the February inventory, we shut down and evacuated the test loop although we do not plan to make this a routine procedure.
5. A task force is already working on modifications to the purge cascade and as their work progresses, the impact on inventory accuracy will also be reviewed.
6. Data was taken during the last inventory to determine the potential effect of transients and further analysis will likely be done.
7. We have started a modest random sampling plan to compare Paducah's weights and analyses with our values.
8. We have done extensive analysis on the variance of our own scales in K-1423.
9. A plan has been developed for determining variance in sampling and we're proceeding on a small scale to better understand the magnitude of the problem and its associated cost.
10. While correcting tare weights on cylinders is pretty much a function of the dollars available, we are looking into what effect incorrect tare weights can have on MUF.
11. Regarding internal volume and surface area calculations, several man-years of effort are being devoted to a complete overhaul of this data. Historically, cascade pressure increases have been accompanied by increases in the measured inventory deficiency and conversely whenever pressures have been lowered inventory has shown an indicated gain (or rather in most cases a decrease in the amount of material that cannot be accounted for). One of the most

likely causes of these variations in calculating inventories is an understatement of volume of cascade equipment. Ray Greene in Don Kellogg's group of the Engineering Division is doing many of these calculations on a time-shared computer system developing routines for T's, junctions, double sweep T's, elbows, bellows, etc., and now is working on an approximate integration technique for evaluating configurations such as surfaces of rotation. As an indication of the size of this project, 1500 drawings are being reviewed and evaluated to complete the basic piping sub-assemblies for the K-33 building alone.

12. We do not have immediate ^{plans} for extensive work on revising inventory equations. I do recognize that we have had only one verification for axial equipment which was made over ten year ago. Our approach here is to try to weigh the cost and potential advantages of going into a more thorough review of these equations.
13. A similar approach is being taken with regard to cut-off times of material in and out of the cascade. A considerable gain can be made here, we feel, through better procedures and perhaps some additional personnel on an as-required basis during the actual taking of the inventory.
14. Our approach to the effects of hidden inventories, UF₅, absorbed UF₆, UO₂F₂, again is to explore the costs and potential pay-off of doing more basic research in this area.
15. We feel that a modest number of experiments, carefully designed, will give us considerably more insight into the contribution of MUF variations due to analytical and instrument errors.
16. Operations Engineering, Statistical Analysis, and Computer Applications and operating groups are on a selective basis resolving problem areas regarding cascade information being used in inventory calculations.. This is a continuing effort by two to three people in conjunction with their other responsibilities.

With regard to long term accountability, plans are being made to evaluate the effect of CIP activities on MUF and develop procedures so that accurate information can be obtained regarding the material which is recovered through decontamination. We are also doing considerable statistical analysis in order to better understand the extent to which the above various areas have contributed to MUF and insure that any gross deviation from past history is immediately recognized and can be effectively evaluated.

A great deal of money could be spent in additional measuring devices, computers, and other hardware, as well as a larger staff, but our approach to date has been one of better utilizing what we have and before we spend additional money insuring ourselves that there will be a potentially significant payoff.

E. H. Krieg, Jr.

EHK:jp

3/12/71